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Comparison of Shuffler and Segmented Gamma Scanner Measurements of 55-Gallon Drums Containing HEU Embedded in Varied Matrices

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Abstract

Many forms of nuclear waste materials are constantly generated at Los Alamos' Chemical and Metallurgical Research (CMR) Facility. For disposal, accounting of the nuclear material must be done. We were recently tasked with measuring sixteen 55-gallon drums, each containing about 150 to 250 grams of HEU. The lower two-thirds of the drums consisted mostly of paper, plastics, and other combustibles. The HEU in this bulk waste had been previously assayed using a segmented gamma scanner (SGS) and ranged from 1 to 20 grams per drum. The drums were later re-opened and specific packages of HEU waste solids, mostly polyethylene bottles filled with uranium oxide and/or fluoride, were placed on top of the bulk waste. The number of bottles ranged from one to seven with an additional ^{235}U mass between 150 to 230 grams per drum. Although many of these additional mass values were based on results from an old chemical analysis, they are expected to be reasonably accurate. We counted the drums on the SGS and shuffler instruments. The results were biased low on the SGS which was calibrated for normal combustible waste (not bottles embedded in waste) and biased high on the shuffler calibrated for small cans of oxide. The biases were caused by the differences among the calibration standards and the drums. This paper is a study of the nature of these biases, providing corrections and comparing the resulting performance characteristics of the instruments. This effort was greatly enhanced by having photographic as well as real-time radiographic information on many of the drums' contents. A new software tool, recently developed at Los Alamos to calculate shuffler count rates accurately and generate bias correction factors, was applied to these drums. The application of the calculational tool to generate bias corrections for these types of matrices is described. The results from the two instruments are compared and discussed.

Introduction

Because of support of multiple research and programmatic activities at Los Alamos, many varied forms of nuclear waste material are constantly being generated at the Chemical and Metallurgical Research (CMR) Facility. This waste must be continually moved along its proper disposition path and out of the facility. To accomplish this, the nuclear material must be properly accounted for in as accurate, efficient, and cost effective manner as practicable. Nondestructive assay (NDA) techniques generally prove best at meeting these objectives. However, it isn't always possible to package the waste in a manner conducive to the most accurate NDA results. In these cases we often have to rely on several different measurement methods and/or develop innovative approaches.

We were recently tasked with measuring sixteen 55-gallon drums, each said to contain about 150 to 250 grams of uranium. The drums originally consisted of low-density combustibles such as paper, rags, and plastics, as well as crushed cans in some of the drums. At this stage they had been assayed with our segmented gamma scanner (SGS) and found to contain 1 to 20 grams of HEU per drum. At a later date it became necessary to open the drums and add additional HEU waste solids to each one. These solids consisted mostly of oxides and/or fluorides packaged in polyethylene bottles or canisters. Ten of the drums had only one canister added whereas the remaining six had anywhere from two to seven. One of the drums even had a uranium fuel element in addition to three canisters.

The matrix material in the bottles/canisters fell into two general categories:

1. Those containing either sodium fluoride (NaF) or activated alumina that was used as a trap material for removal of small quantities of UF_6 from a gas stream/vacuum line. The reaction of the UF_6 with the alumina yields UF_4 plus aluminum fluoride (AlF_3), while the NaF yields a $NaUF_7$ complex.
2. If the bottle/canister did not contain a fluoride based matrix then it most likely contained some form of uranium oxide, either mixed with a variety of materials or coated onto thin pieces of metal. In some cases, graphite may have been mixed with the oxide, however these cases were rare.

All of these waste items were created as part of the research and development, in addition to the HEU processing, which occurred at Los Alamos during the past 20-30 years. The canisters in four of the ten drums with the single add-ons fell into the first category, the remaining six into the second. The multiple canisters in the final six drums were a mixed bag consisting of several different matrix types, with the one aforementioned drum containing the uranium fuel element.

We measured these sixteen drums, with add-on items, on our SGS and then on the shuffler. Using the available calibrations that did not match the drums of interest, the results were biased low on the SGS and high on the shuffler. The intent of this paper is to explore these biases, provide corrections where possible, and compare the performance characteristics of the two instruments. This effort was greatly enhanced by having photographic as well as real-time radiographic information on the drum contents.

SGS Measurement Concerns

As pointed out in the introduction, the results on the sixteen drums, after the additional items were added, were all biased low on the SGS. In order to develop an understanding of the nature of the problem, it is necessary to understand some of the issues surrounding the SGS technique. Because the instrument has been around for several decades now, the literature abounds with SGS measurement limitations.[1,2]

The SGS determines the quantity of nuclear material in a sample by measuring the emitted γ radiation and comparing the corrected count rate with that from known calibration standards. The count rate must be corrected because the γ rays are attenuated as a function of energy and distance through the sample matrix. The correction is determined by using a γ-ray source of known strength (the transmission source) and measuring its absorption through the sample.[3] The amount of absorption is a function of sample density and uniformity. An averaging process, invoked by rotating and translating the sample during the measurement cycle, helps in reducing the effects caused by sample inhomogeneity. Data are therefore acquired for a number of horizontal slices spanning the sample height. These slices are summed at the end of the assay to obtain the total content. If the sample is too dense or nonuniform to allow proper functioning of the transmission source, a bias will be introduced.

A special case of transmission difficulty, known as end or edge effects, occurs during the measurement at the top and bottom of every sample. These effects arise because the transmission measurement does not representatively survey the material that is producing the γ radiation in that area (the radiation cone from the transmission source, due to design limitations of the instrument, does not totally overlap the radiation cone defined by the detector collimation). It is obvious that this effect can also arise due to the varying (always smaller) diameters of the sample if it is offset radially with respect to the calibration standards.[1] These end effects are a special case of the more generalized uncertainties that arise when any inhomogeneities exist within the sample matrix. These effects can extend over a number of segments, depending on the instrument and sample geometry, and the result always biases the final answer low because the transmission source γ rays suffer less attenuation than those from the nuclear material in the matrix resulting in correction factors for the affected segments that are too small. The degree to which this phenomenon affects the final answer is dependent upon the amount of radial offset, the degree of inhomogeneity, and the sample height to collimator ratio; a taller sample has a smaller fraction of affected segments.

Another difficulty arises if the nuclear material, uranium in the present case, is in the form of large particles or lumps. If the lumps are of sufficient radius, then the γ-rays will be attenuated while escaping from the lumps and a low bias will be generated. This phenomenon, known as self-absorption, is completely independent from the aforementioned process of attenuation through the sample matrix. Self-absorption, although addressed in the past,[4] is a difficult SGS measurement issue and has yet to be adequately implemented and field tested.

Because the SGS at the CMR facility is used primarily for assaying low-density combustible bulk waste in 55-gallon drums, the instrument is configured in a large-scale geometry. For example, the detector has a 2-inch collimator and the instrument is calibrated on uranium drum standards not suitable for the small canisters in these drums (approximately 4 to 6 inches in diameter and 8 to 12 inches in height). Also, X-ray imaging revealed that most of the canisters were only about half full and tilted at various angles which make the large-scale geometry configuration even less appropriate. The SGS uranium drum standards[5,1] consist of 20 four-liter bottles about 70% full of a uniform mixture of U_3O_8 and diatomaceous earth. Eighteen bottles are placed around the periphery of the drum, three deep, in six holes cut in the Cello Tex matrix; the final two bottles are placed in a central hole and offset vertically from the outer bottles. The Cello Tex matrix and U_3O_8/diatomaceous earth mixture have the same bulk density (0.26 g/cm^3) and linear attenuation coefficient. The U_3O_8 loading in each bottle is low and does not affect the bulk absorption properties of the standard.

Due to the large detector collimation, which we weren't able to change, and the mismatch between the drum calibration standards and the canisters, a large low bias was inevitable. If all the matrix characteristics (homogeneity, geometry, uniformity of nuclear material distribution, etc.) of the standards, except height, match the assayed items reasonably well, it is possible to manually correct for the end effects bias.[2] Because this was obviously not the situation here, the only recourse was to locate standards that more closely approximate the items. Fortunately, we were able to identify the shuffler uranium oxide standards as viable candidates. These NIST-traceable standards consist of well characterized uranium oxide (U_3O_8) powder enriched to 92.41% in ^{235}U. The standards are packaged in thin-walled, tin-plated steel containers 5 inches in diameter. The different masses give different fill heights (ranging roughly from less than an inch for the 50-gram standard to several inches for the 1000 gram) and, consequently, different ratios of height to diameter for the oxide.[6] The match was fairly close with regard to geometry and matrix density. However, nothing was known about the homogeneity, uniformity of uranium distribution, or uranium particle size of the canister item matrices although there was no a priori reason from the process chemistry to believe the particle sizes would be large enough that self-absorption would be a concern. In any case, we thought it was worth a try and we present the results in the next section.

SGS Measurement Results

Figure 1 shows one of the drums with the top removed. Most of the contents are bulk

Figure 1. Open drum with one add-on at right

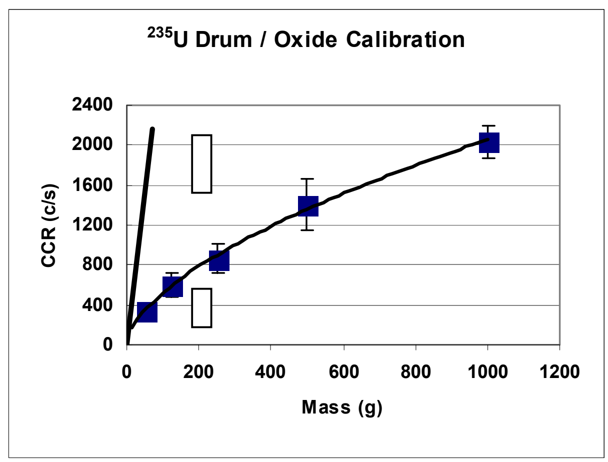

Figure 2. Calibration with data locations

waste items that the SGS is designed to measure as accurately as possible. Close observation reveals an add-on canister located on the right periphery of the drum. The matrix in this canister falls into category number 2 (see Introduction). As already mentioned above, we calibrated the SGS with our shuffler oxide standards to try to more closely represent these two matrix categories. The results are shown in Figure 2 above as the curved line with data points. The straight line curve at the far left is the SGS drum standard calibration. Because those standards are designed to be ideal for low-density bulk waste in 55-gallon drums, edge effects are minimized, transmission is ideal and there are no self-absorption issues. Consequently, the corrected count rate per gram is relatively high at about 30 c/s whereas the average for the shuffler standards is about a factor of 10 lower. Normally the calibration would be linear. However, large edge effects and probable incorrect transmission corrections lead to the nonlinearity.

Figure 2 shows the locations of the measurement results on the two categories of matrices described in the introduction; category 1 items fell above the shuffler standards calibration and category 2 fell below. The six drums with multiple add-on items roughly overlapped the category 1 items and aren't shown for clarity. While the shuffler standards do a better job than the drum standards, as expected, the results are still less than satisfactory.

By simply weighing some of the canisters, the category 2 matrices were almost three times more dense than those of category 1. This is consistent with the uranium oxide being coated onto thin pieces of metal. The transmission source would not be able to correct for attenuation losses through the metal and that would likely account for the low count rate. The much less dense category 1 matrix would be more amenable to attenuation correction and, in fact, is apparently better even than the shuffler standards. The SGS segment data output from the instrument indicated that both matrix categories were being corrected about equally. This, of course, is simply a reflection of the bulk matrix correction and enormous edge effects and not any actual attenuation in the canister matrix.

We really don't have enough information here to say anything definite about possible self-absorption. If it were occurring, it would probably be more likely in category 1 because the category 2 matrix is spread out as oxide on thin metal sheets. This goes counter to the larger count rate per gram in category 1. However, if lots of thin metal pieces are put into a bottle, making a single thick piece, then category 2 could exhibit significant self-absorption. It is interesting to note that far-field measurements, using a single germanium detector and spanning the full angular range of the drums, also reveal huge attenuations but no new information is obtained on the origin of the γ-ray losses.

Shuffler Measurement Results

The shuffler could not be calibrated with physical standards matching the drums with their diverse contents, but the measured count rates could be verified as consistent for the declared contents. Accurate count rates can be calculated,[7,8] assuming the declarations are correct; if the calculated and measured count rates agree, the declarations must be correct with a high degree of confidence.

For each drum the following information was known: (a) weight of the empty drum, (b) gross weight of the drum, (c) gross weight of the can containing most of the uranium, (d) the ^{235}U enrichment, (e) the measured delayed neutron count rate, (f) the flux monitor response (related to the mass of hydrogen in the drum), and (g) the general nature of the matrix in the drum (from visual and X-ray inspection), but not the quantitative amounts of each material in the matrix. Information on the matrices in the various containers bearing the uranium was sparse.

From this information we devised models for the calculations. For drums known to have both hydrogenous materials and crushed metal cans, the ratio of the weights of the two materials was a variable constrained only by estimates from the visual and X-ray inspections. Count rates for most of the drums were calculated and reasonable ratios of the two materials were found by matching the calculated and measured count rates.

As a check on the validity of this process, two drums were opened and the weights of the hydrogenous and metallic materials were measured individually. The declared parameters for these drums were nearly identical but the shuffler's count rates and flux monitor count rates were somewhat different. The drum with the higher flux monitor count rate, i.e. more hydrogen, had the higher shuffler count rate as expected because more moderator generally increases the fission rate and delayed neutron production rate. The unpacking showed that the matrix in the drum with the higher shuffler count rate was entirely hydrogenous (no metal) while the matrix in the other drum was 71% metal by weight. This metal displaced some hydrogenous matrix from the drum, reducing the amount of moderator and the shuffler count rate. Prior to this unpacking, our calculations had told us that there is 73% metal by weight, so the quantitative nature of the matrix as determined by the calculations is quite accurate. The declared parameters for these drums must be accurate for agreement this good.

Since it is impractical to open and weigh the hydrogenous and metallic materials in each drum, reasonable estimates of the relative amount of these quantities in the matrix of each drum were made. For a case in which the measured count rate was 625 counts/s, three iron to polyethylene weight ratios (Fe/CH_2) were modeled: 50/50, 70/30 and 90/10. These models yielded count rates of 1284, 858 and 562, respectively. Given these results, the experimental count rate would be matched for a Fe/CH_2 ratio of about 85/15. Because of the higher density of cans relative to bottles, such a weight ratio is reasonable.

In one case our calculations could not reproduce the measured count rate using reasonable ratios of hydrogenous and metallic matrices. But the matrix in the uranium-bearing can is apparently alumina, a material that can absorb moisture from the air. Even when the trapped water is only 10% of the alumina by weight, the count rate is greatly enhanced by this moderator in close proximity to the uranium. This is a plausible resolution of the difficulty with the calculation in this case when using dry alumina.

Two other drums with 100% hydrogenous matrices had measured count rates much smaller than the calculated rates. No neutron absorbers (such as boron) are thought to be in the drums, but the exact natures of the matrices in the containers is poorly known. We cannot account for the discrepancies in these two cases; the mostly likely cause of the high calculated rates is with the description used for the matrices in the containers.

In general, the declared parameters along with plausible assumptions for these drums were found to lead to calculated count rates consistent with the measured shuffler count rates. This is a verification of the declared parameters.

Summary and Conclusions

We studied waste 55-gallon drums that contained very localized concentrations of approximately 200 grams of uranium amid mostly hydrogenous matrices with about 10 grams of uranium. This means a drum had most of the uranium in a localized inhomogeneity and the rest was spread within a nearly homogenous drum. Not surprisingly, these drums were found to be difficult to assay; attempts were made with both a shuffler and an SGS. Earlier far-field measurements demonstrated that gamma-ray attenuations were severe. The existing calibrations for all these instruments were for radically different materials and naturally gave strong biases. Corrections for these biases were attempted for the shuffler and SGS.

A computer model of the shuffler with a waste drum was used to calculate count rates that in effect included the bias correction for the available but inappropriate calibration. With assumptions about the nature of the matrices based on known drum weights, X-ray examinations, and detailed weights of hydrogenous and metal matrix materials in two drums, we could show that the measured count rates were plausible, aside from two cases where calculated rates were higher than measured rates.

The biases in the SGS results consistently made the measured results lower than the declared masses. But having almost all the uranium concentrated in a can or bottle on the edge of a drum violates an assumption of the SGS that a segment of the drum is homogeneous. Appropriate attenuation corrections were impossible to find under this condition. The gamma-ray attenuation by materials within a container would then be uncorrected, giving measured masses biased low. A new calibration was generated using available cans of uranium oxide but the measured masses were not close to this calibration either because the real cans and bottles had matrix materials that the calibration cans did not. For example, some of the cans and bottles were filled with small metal pieces, unlike the cans of pure oxide.

Better bias corrections could be made for the shuffler technique only after more effort is put into discovering the materials in the matrices in the drum and particularly inside the cans and bottles containing most of the uranium. The SGS needs physical standards that better match the real drums, but even then the assumption of homogeneity within a segment is violated and transmission corrections would not be made correctly. Far-field gamma-ray measurements also need transmission corrections to account for the variety of matrix materials inside the cans and bottles as well as the drum.

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